Magnetic Diode at $T = 300$ K

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We report the finding of unidirectional electronic properties, analogous to a semiconductor diode, in two-dimensional artificial permalloy honeycomb lattice of ultra-small bond, with a typical length of $\simeq$ 12 nm. The unidirectional transport behavior, characterized by the asymmetric colossal enhancement in differential conductivity at a modest current application of $\simeq$10-15 μA, persists to $T = 300$ K in honeycomb lattice of thickness $\simeq$ 6 nm. The asymmetric behavior arises without the application of magnetic field. A qualitative analysis of experimental data suggests the role of magnetic charge or monopoles in the unusual observations with strong implication for spintronics.

Semiconductor diode and transistor are key elements in modern electronic devices. The substitution of a semiconductor by a magnetic system with resembling properties, such as an unidirectional biasing of electric current, is expected to enhance the functionalities of the device significantly. It can play the dual role of the reprogrammable logic and the non-volatile memory element.[1–3] Multilayered magnetic devices are known to exhibit a very high level of unidirectional spin polarization in applied magnetic field, which can be utilized to create a magnetic diode with unidirectional current biasing properties.[2] More recently, synergistic experimental and theoretical research efforts on the development of a magnetic diode have led to the development of new spintronics devices that includes the design of a nanomagnetic logic device, bipolar spin diodes and the unipolar spin diode across a magnetic domain wall.[3–8] However, an application of magnetic field is required in most cases. A magnetic system, which can exhibit unidirectional current biasing at a modest current (resulting in reasonably small output power) without the application of magnetic field, is still elusive. Additionally, any such device must also demonstrate the operational ability at room temperature for practical applications.

In this letter, we report the observation of unidirectional current biasing, analogous to a semiconductor diode, in the electronic measurements on artificial permalloy honeycomb lattice. The unidirectional electronic properties arise without the necessity of magnetic field application. The phenomenological observation of diode-type function in artificial honeycomb lattice possibly hints of new and unexplored properties of magnetic charges (or monopoles) that are argued to exist in this system.[9, 10] The design of two-dimensional artificial honeycomb lattice was originally envisaged to complement the study of spin ice phenomenon and associated magnetic monopoles in three dimensional geometrically frustrated magnets.[9, 11] Since then, it has evolved in a new research arena to explore novel magnetic and electronic properties.[12–14] For a moderate aspect ratio, defined by l (length) $/t$ (thickness), of the connecting element of the honeycomb, the magnetic moment lies along the length of the element due to magnetostatic interaction and shape anisotropy.[10, 15] Consequently, two types of local spin configurations emerge: all moments either point to or away from a vertex of the honeycomb, also called 'all-in or all-out' configuration or, two moments pointing in and one point away from the vertex (or vice-versa), also called 'two-in and one-out' (or vice-versa) configuration.[10, 16] The latter spin arrangement is also termed as the spin-ice configuration, which results in a net magnetic charge of ±1 unit to a given vertex. We have created new artificial honeycomb lattice of connecting ultra-small bonds with a typical size of 12 nm in length and approximately 5 nm in width, see Fig. 1a-b. Using detailed magnetic, electrical and neutron scattering measurements on a macroscopic size sample of thin ($t\simeq$5 nm) artificial honeycomb lattice, previously we demonstrated the temperature dependent evolution of theoretically predicted novel magnetism that includes a paramagnetic spin gas at high temperature followed by the development of short-range ordered spin ice state at intermediate temperature, 30 K $< T <$220 K - 250 K, and finally a tendency to develop the spin solid state, manifested by the distribution of chiral vortex states, below $T = 30$ K.[12, 13, 17]

Measurements of differential conductivity on newly fabricated artificial honeycomb lattice of varying thicknesses have revealed electronic properties that are reminiscent of a semiconductor diode. It is found that the differential conductivity increases by more than two orders of magnitude for a modest unidirectional current application of $\simeq$ 10 μA, compared to the negligible value near zero bias current. Electronic measurement for the oppositely directed current yields a very small or negligible differential conductivity. The temperature dependence of unidirectional biasing strongly depends on the thickness of the lattice. For instance, in a very thin lattice, $t \simeq 2$ nm, the unidirectional biasing is nearly absent. As the thickness of the honeycomb lattice increases to $t \simeq 4$ nm, this phenomenon starts developing as temperature increases and becomes very pronounced in a temperature range of 30 K $< T <$250 K; incidentally, the same temperature regime where the short ranged ordered spin ice

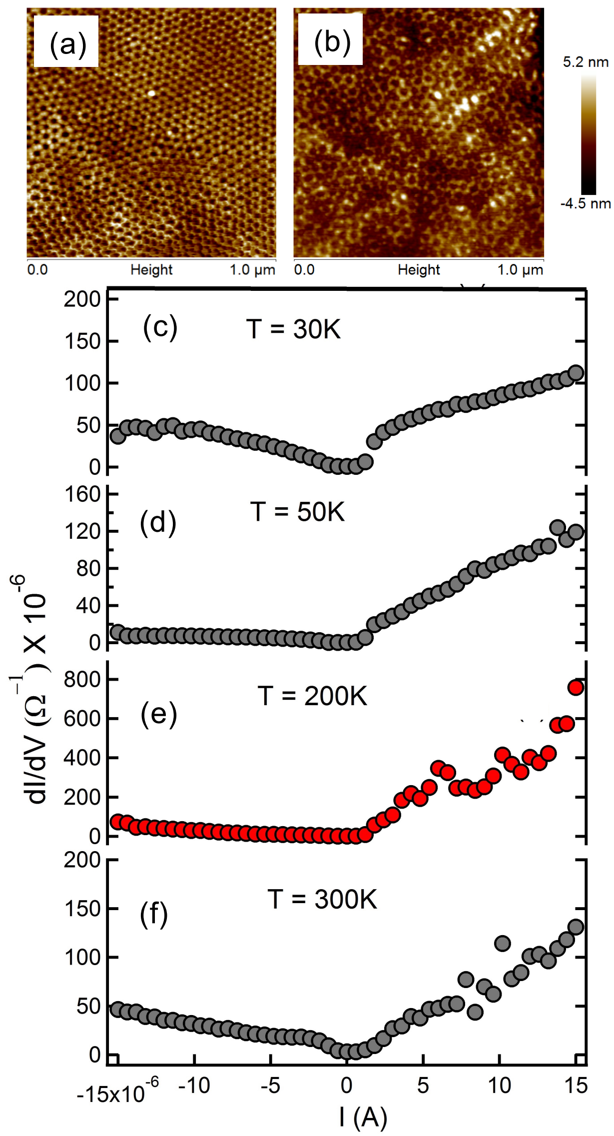

FIG. 1: Atomic force micrographs of artificial honeycomb lattice and electronic properties measurement data. (a-b) Atomic force micrographs of typical artificial honeycomb lattice, after reactive ion etching and material deposition. (c-f) Plot of $d\mathrm{I}/d\mathrm{V}$ as a function of current bias, obtained on a honeycomb lattice of thickness $t \simeq 4$ nm, at few characteristic temperatures. A colossal enhancement in differential conductivity at $I \simeq 10$ μA compared to the zero bias value, is observed at $T = 30$ K. As temperature increases, the enhancement in differential conductivity becomes highly asymmetric, suggesting a semiconductor diode-type unidirectional electronic properties. ($d\mathrm{I}/d\mathrm{V}$) tends to become symmetric again as $T \to 300$ K, albeit weaker.

state was reported to exist in a honeycomb lattice of similar thickness.[17] At the lattice thickness of $t \simeq 6$ nm, the asymmetric behavior is found to persist to $T = 300$ K. For further increase in the thickness of the lattice, unidirectional conductance gradually weaken and becomes indistinguishable from the flat background conductivity. Qualitative analysis of the experimental data suggests the possible role of inter-elemental energy behind the unusual observations in artificial honeycomb lattice.

Artificial honeycomb lattice of ultra-small bond was fabricated using a combination of diblock copolymer template synthesis, reactive ion etching and material deposition at near parallel angle to achieve the two-dimensional characteristic of the system. For the synthesis of diblock template, we utilized a diblock copolymer polystyrene-b-poly-4-vinyl pyridine (PS-b-P4VP) of molecular weight 23 K Dalton. It generates a large uniform hexagonal nanoporous template with the pore dimeter of $\simeq 12$ nm and a lattice spacing of $\simeq 27$ nm. Details about the synthesis of diblock template can be found somewhere else.[18] Next, the diblock template was used as the etching mask to transfer the hexagonal pattern to the underlying silicon substrate using CF_4-based reactive ion etching technique. A thin layer of diblock porous template, 4-5 nm thick, still remained present on top of silicon substrate. The top layer of the substrate resembled a honeycomb lattice pattern. This topographical property was exploited to create metallic honeycomb lattice by depositing permalloy, $Ni_{0.8}Fe_{0.2}$, in near parallel configuration (within 1^o degree) using a new sample holder, specifically designed to limit the deposition to the top of the substrate only, in an electron-beam evaporator. The substrate was rotated at a moderate constant speed about its axis during the deposition process to create uniformity in the film thickness. Atomic force micrographs of typical artificial honeycomb lattice, fabricated using technique, are shown in Fig. 1a-b. Electronic measurements were performed using a synchronized combination of Keithley current source meter 6221 and a nanovoltmeter 2182A, via a trigger link, on $\simeq 8$ mm $\times$ 6 mm size samples. For the measurement purposes, electrical contacts were made in the four-probe configuration using commercially available silver paint. Electronic measurements involved the averaging of twenty data points where each data was filtered for 20 seconds before recording the final value. Samples were always stored in a vacuum container.

In Fig. 1c-f, we plot differential conductivity, $d\mathrm{I}/d\mathrm{V}$, results on a honeycomb lattice of thickness $t \simeq 4$ nm as a function of applied current bias at few characteristic temperatures. Two features are immediately observed in the differential conductivity plot at $T = 30$ K: first, $d\mathrm{I}/d\mathrm{V}$ increases by more than two orders of magnitude with respect to the zero-bias value at the current application of $\simeq 10$ μA and second, the colossal gain in differential conductivity starts developing asymmetric character with respect to the current bias direction. The asymmetric behavior becomes prevalent as temperature increases above $T = 30$ K. As shown in Fig. 1d, the measurement at $T = 50$ K manifest drastically different electronic responses to oppositely directed current biases. Compared to the very small change in $d\mathrm{I}/d\mathrm{V}$ for negative current bias, a colossal enhancement in differential conductivity is detected for positive current bias. It indicates an unidi-

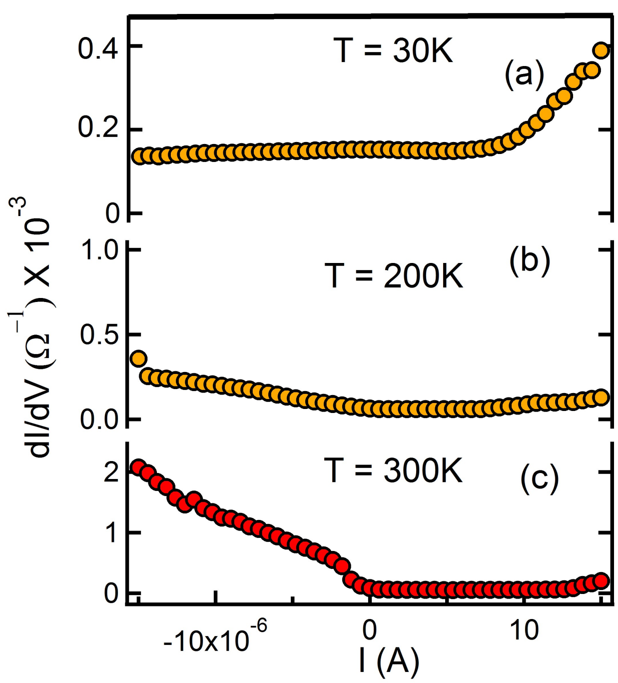

FIG. 2: Persistence of diode-type electronic behavior to room temperature in $t \simeq 6$ nm thick artificial honeycomb lattice. (a) Measurement of differential conductivity in honeycomb lattice exhibit unidirectional electronic transport to $T = 30$ K. (b) As temperature increases to $T = 200$ K, the asymmetric behavior weaken. (c) Interestingly enough, the unidirectional electronic properties persists to $T = 300$ K; albeit with opposite current bias polarity.

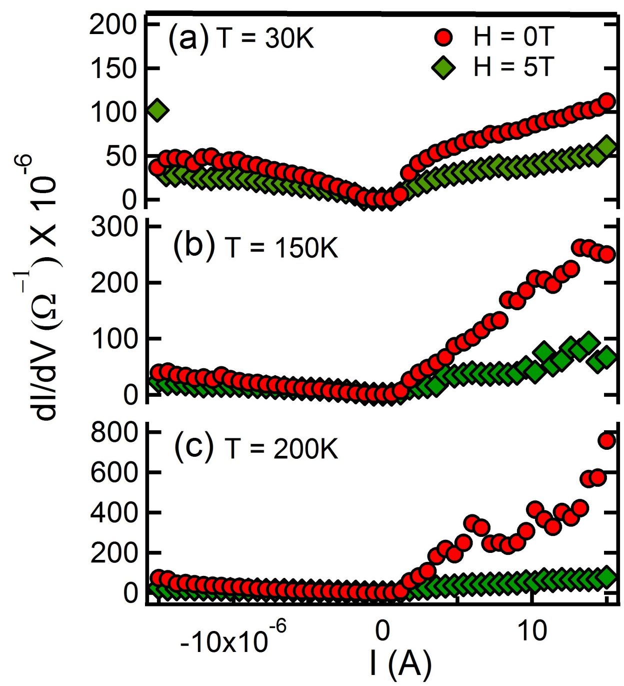

FIG. 3: Magnetic field effect on electronic properties. Here we show experimental results on the honeycomb lattice of thickness $t \simeq 4$ nm, as it exhibits both symmetric and asymmetric current biasing as a function of temperature. (a) At $H = 5$ T, $d\mathrm{I}/d\mathrm{V}$ decreases at high current bias at $T = 30$ K. Yet, it maintains the near-symmetric character. (b-c) The field effect is very pronounced at higher temperatures. A field application of $H = 5$ T strongly suppresses the asymmetric nature of electronic transport and becomes almost symmetric.

rectional electronic transport in the artificial honeycomb lattice of ultra-small bond, which becomes significantly pronounced as temperature increases to $T = 200$ K (see Fig. 1e). Such an unidirectional electronic transport is the hallmark of a nonmagnetic semiconductor diode. Electronic measurements were also performed for reverse current sweep to check for possible hysteresis in the conductivity data. But no such behavior was detected. The asymmetric behavior tend to become symmetric again, albeit weakly, as $T \rightarrow 300$ K.

Increasing the thickness of the honeycomb lattice profoundly impacts the temperature dependence of the unidirectional current biasing. A detailed study was carried out in this regard by systematically varying the thickness of the lattice between $\simeq 2$ nm to 9 nm. Differential conductivity data, obtained on honeycomb lattice of varying thicknesses, are shown in Fig. S1 in the Supplementary Materials. It is found that increasing the thickness of the lattice progresses the onset of the unidirectional electronic transport to lower temperature in the system. Also, no asymmetric enhancement in differential conductivity is observed at any temperature in very thin, $\simeq 2$ nm, honeycomb lattice. Most surprising behavior, however, is observed in honeycomb sample of intermediate thickness. As shown in Fig. 2, electronic measurements on $t \simeq 6$ nm thick artificial honeycomb lattice depict the persistence of strong unidirectional transport to $T = 300$ K, for oppositely directed current compared to $t \simeq 4$ nm thick sample. Interestingly, the system tends to preserve the unidirectional conductivity enhancement even at $T = 30$ K. However, the current bias direction is reversed between $T = 300$ K and 30 K. The changeover occurs around $T = 200$ K where very little change in differential conductivity, with respect to the near zero bias value, is observed. Measurements were repeated multiple times and on multiple samples to verify the reproducibility of unidirectional electronic transport in artificial honeycomb lattice. The asymmetric current biasing gradually disappears as thickness of the lattice increases above $t \geq 8$ nm (see Fig. S1 in the Supplementary Materials).

Next, we have investigated magnetic field effect on the electronic properties of artificial honeycomb lattice. Magnetic field was applied in-plane to the sample. In Fig. 3, we plot the differential conductivity data, obtained on a honeycomb sample of characteristic thickness $t \simeq 4$ nm, in applied fields of $H = 0$ and 5 T. At a field application of $H = 5$ T at $T = 30$ K, $d\mathrm{I}/d\mathrm{V}$ decreases significantly at higher current bias, compared to the zero field value. Yet, the symmetric nature of conductivity biasing is maintained. A more dramatic field effect is ob-

served at intermediate temperature, 30 K $< T <$ 250 K, where the system tends to exhibit strong unidirectional electronic transport in zero field. Measurement at $H =$ 5T at $T = 150$ K show that the asymmetric behavior is strongly suppressed and differential conductivity tends to become symmetric. A similar behavior is observed in the electronic measurements at $T = 200$ K. Similar field dependence of differential conductivity are observed in other samples. While the field dependence of $d\mathrm{I}/d\mathrm{V}$ confirms the role of magnetism in the electronics transport measurements, a lower conductivity in applied field is somewhat surprising. At such a high field, magnetic moment will tend to align along the field direction and thus creates an effective ferromagnetic (FM) type structure. In general, a ferromagnetic configuration gives rise to lower resistance (or higher conductivity) in electronic measurements.[19] Further research works, both theoretical and experimental, are desirable to understand the field dependence of electronic properties in this system.

By drawing analogy with the current carrying characteristic of a p-n junction semiconductor diode where the current is controlled to flow in the desired direction by using p- or n-type dopants in a semiconductor, it can be argued that magnetic equivalent of electric charge carriers (electron and hole) must exist. Previously, we demonstrated the evolution of various magnetic phases as a function of temperature in an artificial honeycomb lattice ($t \simeq 5$ nm). It was found that the honeycomb lattice undergoes a transition from paramagnetic or spin gas phase to the short-range ordered spin ice phase below $T \simeq 250$ K. At low temperature, $T \leq 30$ K, the system tends to develop a spin solid state, manifested by the distribution of chiral vortex states of opposite polarities. The development of spin solid state becomes pronounced as $T \rightarrow$ 0 K.[17] In the spin solid state at low temperature, the vortex circulation of magnetization does not exhibit any preferential direction. Incidentally, there is an one-to-one correspondence between the occurrence of unidirectional electronic transport, in a similar honeycomb lattice ($t \simeq$ 4 nm), and the spin ice phase. Since conduction electrons follow the magnetization direction,[20] the nearly symmetric (or weakly asymmetric) colossal enhancement in differential conductivity at low temperature ($T \leq 30$ K) is not surprising. At $T \leq 30$ K, the magnetization configuration mostly consists of the spin solid state, as reported previously, with spin ice state as the minority contributing phase. Consequently, electronic measurement at $T = 30$ K depicts a nearly symmetric conductance. On the other hand, at intermediate temperature, 30 K $< T <$ 250 K, the short-range ordered spin ice state can develop preferential conductivity biasing along the local anisotropic direction. Previous research works on artificial honeycomb lattice have demonstrated the existence of magnetic monopoles, or magnetic charges, in the spin ice state.[9, 21] This could be a possible reason for the unidirectional current biasing in the newly fabricated artificial honeycomb lattice.

Our qualitative explanation of the unidirectional electronic transport in honeycomb lattice does not explain the persistence of asymmetric biasing to low temperature in $t \simeq 6$ nm thick honeycomb lattice or, the absence of this unusual behavior in very thin, $t \simeq 2$ nm, lattice (see Fig. S1). It can be argued that as the honeycomb lattice becomes thicker, the gain in the inter-elemental energy prohibits the development of the zero entropy density spin solid state. Therefore, the system remains locked in the spin ice state even at low temperature and exhibits the asymmetric current biasing. However, the same sample exhibits a reversal in the unidirectional transport property as temperature reduces form T = 300 K to 30 K. Similarly, a thinner honeycomb lattice corresponds to a smaller inter-elemental energy. Hence, the onset temperature to the spin ice phase will be reduced in this sample. However, no evidence of unidirectional transport is detected at any temperature. Further research works are needed to fully understand it.

In essence, electronic measurements on newly fabricated artificial honeycomb lattice have revealed interesting, yet highly anomalous, properties with strong implication for practical applications. It is found that the unidirectional colossal enhancement in differential conductivity can persist to room temperature at the right thickness of the artificial honeycomb lattice. Most importantly, the initiation of unidirectional electronic transport does not require magnetic field application. These two properties, the persistence of unidirectional transport to $T = 300$ K for a modest current application (resulting in small output power) and the absence of the necessity of magnetic field application, makes it highly attractive for practical application in spintronics devices.

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